Spin-subband populations and spin polarization of quasi two-dimensional carriers under in-plane magnetic field

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Under an in-plane magnetic field, the density of states of quasi two-dimensional carriers deviates from the occasionally stereotypic step-like form both quantitatively and qualitatively. For the first time in the literature as far as we know, we study how this affects the spin-subband populations and the spin-polarization as functions of the temperature, T, and the in-plane magnetic field, B, for narrow to wide dilute-magnetic-semiconductor quantum wells. We examine a wide range of material and structural parameters, focusing on the quantum well width, the magnitude of the spin-spin exchange interaction, and the sheet carrier concentration. Generally, increasing T, the carrier spin-splitting, $U_{o\sigma}$, decreases, augmenting the influence of the "minority"-spin carriers. Increasing B, $U_{o\sigma}$ increases and accordingly carriers populate "majority"-spin subbands while they abandon "minority"-spin subbands. Furthermore, in line with the density of states modification, all energetically higher subbands become gradually depopulated. We also indicate the ranges where the system is completely spin-polarized.

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I. PREAMBLE

An in-plane magnetic field applied to a quasi two-dimensional system distorts the equal-energy surfaces^{1,2} or equivalently the density of states^{3,4} (DOS). An interplay between spatial and magnetic confinement is established and properly- it is necessary to compute self-consistently the energy dispersion, $E_{i,\sigma}(k_x)$, where i is the subband index, σ denotes the spin, k_x is the in-plane wave vector perpendicular to the in-plane magnetic field, B (applied along y), and z is the growth axis. Hence, the envelope function along z depends on k_x i.e., $\psi_{i,\sigma,k_x,k_y}(\mathbf{r}) \propto \zeta_{i,\sigma,k_x}(z) \exp(ik_x x) \exp(ik_y y)$. This modification has been realized in magnetotransport⁵ and photoluminescence⁶ experiments. An impressive fluctuation of the in-plane magnetization in dilute-magnetic-semiconductor (DMS) structures in cases of strong competition between spatial and magnetic confinement has been predicted at low enough temperatures⁷ and a compact DOS formula holding for any type of interplay between spatial and magnetic confinement already exists⁷.

Although this DOS modification can be extremely significant both quantitatively and qualitatively it is sometimes neglected without a second thought. Naturally, in the limit of very narrow quantum wells (QWs) or for $B \to 0$, the DOS preserves the ideal step-like form. The "opposite" asymptotic limit is a simple saddle point, where the DOS diverges logarithmically³. However, generally, the van Hove singularities which show up are not simple saddle points⁴. Summarizing, models which ignore the above DOS modifications can only be applied to very narrow QWs or for $B \to 0$.

During the last years, the progress in growth, characterization and understanding of transition-metal-doped semiconductors has been impressive^{8,9,10}. As a result, new phenomena have been discovered, e.g. tunnel magnetoresistance, spin-dependent scattering, interlayer coupling due to carrier polarization, electrical electron and hole spin injection, and electric field control of ferromagnetism^{8,9}. Usually the host material is a III-V semiconductor^{8,9,10}. For example, in (Ga,Mn)As or in (In,Mn)As, Mn substitutes a small fraction of cations providing holes and local magnetic moments. Hence, the corresponding structures utilize the valence band. The highest ferromagnetic transition temperature, T_C , reported so far for III-V-based valence-band magnetic semiconductors is 173 K in (Ga,Mn)As epilayers¹⁰.

In II-VI materials, Mn provides only local magnetic moments. The corresponding heterostructures, for example $ZnSe/Zn_{1-x-y}Cd_xMn_ySe$, utilize either the conduction or the valence band, depending on the type of dopants used in the barriers, namely, donors (e.g. Cl, I) or acceptors (e.g. Li), respectively. In the present article we investigate such a system where either the conduction band or the valence band can be exploited for spintronic applications. The key material of each structure (e.g. ZnSe, CdTe etc) may possess quite different material parameters e.g. positive or negative g factors¹¹. We also note that the band gap of common II-VI crystals covers all the range from the

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ultraviolet to the infrared¹⁶. Interestingly the existence of ferromagnetic order in n-doped (Cd,Mn)Te based structures -at extremely low temperatures- has been suggested both experimentally and theoretically^{17,18}.

In a recent publication⁷ we restricted ourselves to DMS structures utilizing the conduction band and to very low temperatures. We studied the spin-subband populations, the internal and free energy, the Shannon entropy, and the in-plane magnetization M as functions of the in-plane magnetic field, for different degrees of spatial confinement. The enhanced electron spin-splitting $U_{o\sigma}$ can be considered as the sum of two terms, α and β . α is proportional to the cyclotron gap, $\hbar\omega_c$, while β arises from the exchange interaction between the itinerant carrier (conduction electron in Ref.⁷) and the localized spins (Mn⁺² cations in Ref.⁷). Notice that in such an approximation the direct exchange interaction between the neighboring localized impurity spins is neglected, being much smaller than the interaction between impurity spins and carrier spins¹⁹, although according to a recent report it might influence the carrier spin polarization²⁰. The very low T impelled us to a drastic first approximation⁷, i.e. to take into account only β , and moreover to approximate the corresponding Brillouin function by 1.

In the present article we attempt a major improvement. Namely, we examine the relative influence of α and β in a wide temperature band (0 to 400 K) and in a wide in-plane magnetic field band (0 to 20 T), as well as in a wide range of material parameters, not necessarily restricting ourselves in the conduction band²¹. Our purpose is to systematically study the influence of the DOS modification on the spin-subband populations and the spin-polarization of quasi twodimensional carriers, as functions of the in-plane magnetic field and the temperature. Besides, we indicate the ranges where the system is completely spin-polarized. In Section II we introduce our theoretical framework. In Section III we examine the spin-subband populations and the spin polarization, ζ , of non-magnetic-semiconductor (NMS) / narrow to wide dilute-magnetic-semiconductor (DMS) / NMS quantum wells (QWs), as a function of the temperature, T, and the in-plane magnetic field, B. We notice that in the present system due to the influence of carriers, increase of the QW width transforms the heterostructure from an "almost perfect square QW" to a "double QW with a soft barrier" ("a system of two separated heterojunctions")²³. Thus, the present heterostructure allows us to study "single" as well as "double" QWs. To facilitate the reader, we provide in Fig. 1 sketches of the self-consistent QW profiles for T=20 K, B = 0 T and sheet carrier concentration $N_s = 1.566 \times 10^{11}$ cm⁻², for QW widths 10 nm, 30 nm and 60 nm. We examine how the DOS modification affects ζ for a wide range of material and structural parameters focusing on the quantum well width, the magnitude of the spin-spin exchange interaction coupling strength, and the sheet carrier concentration. Finally, in Section IV we briefly state our conclusions.

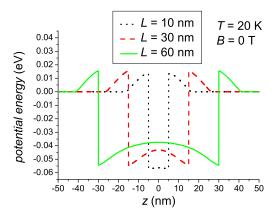


FIG. 1: (Color online) Sketch of the QW profiles for T=20 K, B=0 T and $N_s=1.566\times 10^{11}$ cm⁻², for QW widths 10 nm, 30 nm and 60 nm.

II. THEORY

Under a magnetic field, B, applied parallel to the interfaces, the equal energy surfaces are gradually distorted. The density of states deviates from the ideal step-like form both quantitatively and qualitatively⁷, i.e. it takes the form:

$$\rho(\mathcal{E}) = \frac{A\sqrt{2m^*}}{4\pi^2\hbar} \sum_{i,\sigma} \int_{-\infty}^{+\infty} dk_x \frac{\Theta(\mathcal{E} - E_{i,\sigma}(k_x))}{\sqrt{\mathcal{E} - E_{i,\sigma}(k_x)}},\tag{1}$$

where it is implied that the QW is along the z axis and the magnetic field is applied along the y axis. Θ is the step function, A is the xy area of the structure, m^* is the effective mass²⁴. $E_{i,\sigma}(k_x)$ are the spin-dependent xz-plane eigenenergies. Generally, $E_{i,\sigma}(k_x)$ must be self-consistently calculated^{1,2,4,5,6,7}. Equation (1) is valid for any type of interplay between spatial and magnetic confinement. The k_x dependence in Eq. (1) increases the numerical cost by a factor of $10^2 - 10^3$ in many cases. This k_x dependence is quite often "conveniently" ignored, although this is only justified for narrow QWs. However, with the existing computing power, such a "simplification" is not any more necessary. Only in the limit $B \to 0$, the DOS retains the occasionally stereotypic staircase shape with the ideal step $\frac{1}{2} \frac{m^* A}{\pi \hbar^2}$ for each spin. The opposite asymptotic limit of Eq. (1) is that of a simple saddle point, where the DOS diverges logarithmically³. The DOS modification significantly affects the physical properties^{1,2,3,4,5,6,7}. For completeness, we notice that Eq. (1) ignores the effect of disorder which -with the current epitaxial techniques- is important when the concentration of magnetic ions is high^{26,27}. Disorder will certainly induce some broadening of the spin-subbands.

In DMS structures, the electron spin-splitting, $U_{o\sigma}$, is not proportional to the cyclotron gap, $\hbar\omega_c$, i.e. it acquires the form^{28,29,30}:

$$U_{o\sigma} = \frac{g^* m^*}{2m_e} \hbar \omega_c - y N_0 J_{sp-d} SB_S(\xi) = \alpha + \beta.$$
 (2)

 $\alpha = \alpha(B)$ describes the Zeeman coupling between the spin of the itinerant carrier and the magnetic field, while $\beta = \beta(B,T)$ expresses the exchange interaction between the spins of the Mn⁺² cations and the spin of the itinerant carrier (initially supposed to be an electron). g^* is the g-factor¹¹ of the itinerant carrier. y is the molecular fraction of Mn. N_0 is the three-dimensional (volume) concentration of cations. J_{sp-d} is the coupling strength due to the spin-spin exchange interaction between the d electrons of the Mn⁺² cations and the s- or p-band electrons, and it is negative for conduction band electrons. The factor $SB_S(\xi)$ represents the spin polarization of the Mn⁺² cations. The spin of the Mn⁺² cation is S = 5/2. $B_S(\xi)$ is the standard Brillouin function, while^{19,29}

$$\xi = \frac{g_{Mn}\mu_B SB - J_{sp-d} S \frac{n_{down} - n_{up}}{2}}{k_B T}.$$
(3)

 k_B is the Boltzmann constant. μ_B is the Bohr magneton. g_{Mn} is the g factor of $\mathrm{Mn^{31}}$. n_{down} and n_{up} are the spin-down and spin-up three-dimensional (volume) concentrations measured e.g. in $\mathrm{cm^{-3}}$, while $N_{s,down}$ and $N_{s,up}$ used below are the spin-down and spin-up two-dimensional (sheet) concentrations measured e.g. in $\mathrm{cm^{-2}}$. In Eq. 3 (and only there) we approximate $n_{down} - n_{up} \approx (N_{s,down} - N_{s,up})/L$, where L is the QW width. The first term in the numerator of Eq. 3 represents the contribution of the Zeeman coupling between the localized spin and the magnetic field. The second term in the numerator of Eq. 3 (sometimes called "feedback mechanism") represents the kinetic exchange contribution which -in principle- can induce spontaneous spin-polarization i.e. in the absence of an external magnetic field²⁹. Notice that $n_{down} - n_{up}$ is positive for conduction band electrons. Finally, for conduction band electrons, the spin polarization can be defined by

$$\zeta = \frac{N_{s,down} - N_{s,up}}{N_s}.\tag{4}$$

 $N_s = N_{s,down} + N_{s,up}$ is the free carrier two-dimensional (sheet) concentration.

The use of such a simplified Brillouin-function approach is quite common when dealing with quasi two-dimensional systems^{17,18,28,29,30}. This way, the spin-orbit coupling is not taken into account. This is certainly a simplification, since increasing temperature, the magnetization of the magnetic ions competes with spin-orbit coupling. The spin-orbit coupling^{26,27} induces temperature dependent spin relaxation. Therefore, the carriers' spin-polarization does not only depend on the magnetic order of the magnetic ions, expressed here with the help of the Brillouin function and the carriers' spin relaxation influences the magnetic order of the localized magnetic moments.

The variation of the temperature, T, affects the spin polarization. The spin polarization is also influenced by the magnetic field, in an opposite manner i.e. B tends to align the spins. Furthermore, for each type of spin population, the in-plane magnetic field -via the distortion of the DOS- redistributes the electrons between the subbands. Consequently, the spin polarization can be tuned by varying the temperature and the magnetic field. Indeed, preliminary conduction band calculations for specific values of the material parameters, for very narrow quantum wells, have shown³³ that when the "feedback mechanism" due to the difference between the populations of the spin down and the spin up electrons can be neglected, the spin polarization vanishes for $B \to 0$. The analysis presented above can be useful for p-doped structures, assuming -as usual- that a single valence band description is a fair first approximation.

III. RESULTS AND DISCUSSION

Initially we consider heterostructures of the type n-doped ZnSe / Zn_{1-x-y}Cd_xMn_ySe /n-doped ZnSe. Let us take $y=0.035, -yN_0J_{sp-d}=0.13$ Hartree*, and the conduction band offset, $\Delta U_{cb}=1$ Hartree*²⁸. We notice that for ZnSe, 1 Hartree* ≈ 70.5 meV. ZnSe has a sphalerite-type structure and the lattice constant is ~ 0.567 nm. Hence, $-J_{sp-d}\approx 12\times 10^{-3}$ eV nm³. This is one order of magnitude smaller than the value commonly used for the III-V Ga(Mn)As valence band system $(J_{pd}=15\times 10^{-2} \text{ eV nm}^3)^{19,29,30}$.

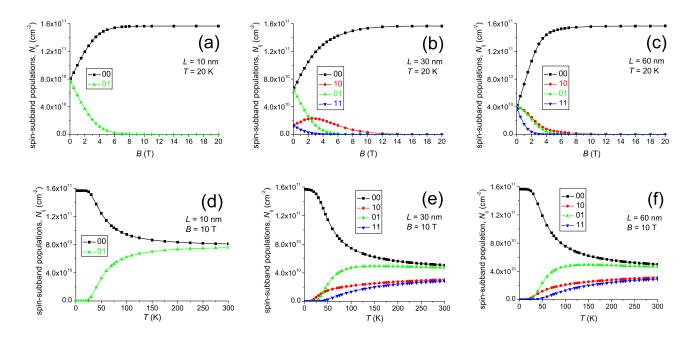


FIG. 2: (Color online) The spin-subband populations $N_{ij} = N_{ij}(B)$ for T = 20 K (first row), and $N_{ij} = N_{ij}(T)$ for B = 10 T (second row) of a n-doped ZnSe / Zn_{1-x-y}Cd_xMn_ySe /n-doped ZnSe QW with y = 0.035. $-J_{sp-d} = 12 \times 10^{-3}$ eV nm³. 00 stands for the ground-state spin-down-subband, 10 for the 1st excited spin-down-subband, 01 for the ground-state spin-up-subband, and 11 represents the 1st excited spin-up-subband. Each column corresponds to different well width i.e. L = 10 nm, 30 nm, and 60 nm.

Figure 2 depicts the spin-subband populations, N_{ij} as a function of B (a-c), and as a function of T (d-f), for three different well widths, namely (a,d) for L=10 nm (b,e) for L=30 nm, and (c,f) for L=60 nm. Initially, we deliberately keep the total sheet carrier concentration constant ($N_s=1.566\times 10^{11}$ cm⁻²), assuming that all dopants are ionized. In (a-c) T=20 K. In (d-f) B=10 T. The pair ij is defined in the following manner: 00 symbolizes the ground-state spin-down-subband, 10 the 1st excited spin-down-subband, 01 the ground-state spin-up-subband, and finally 11 symbolizes the 1st excited spin-up-subband. Due to the small value of J_{sp-d} , the influence of the "feed-back mechanism" due to the difference between spin-down and spin-up concentrations is negligible in the present system. Indeed, since $-J_{sp-d}\frac{n_{down}-n_{up}}{2}$ is negligible here, then for B=0, it follows that (a) $\xi\approx 0$, thus $B_S(\xi)\approx 0$, therefore $\beta\approx 0$, and (b) $g^*\mu_B B=\alpha=0$. Hence, $U_{o\sigma}\approx 0$, and consequently $\zeta\approx 0$. In fact, inspection of Figs.2 (a-c), reveals that for B=0, in Fig.2(a) $N_{00}=N_{01}$, in Fig.2(b) $N_{00}=N_{01}$ and $N_{10}=N_{11}$, and in Fig.2(c) $N_{00}=N_{01}$ and $N_{10}=N_{11}$. For the very wide quantum well (L=60 nm), as expected²³, the four spin-subbands are almost equally populated for B=0. Increasing B, we observe that there are two mechanisms which cause depopulations: (I) The increase of $U_{o\sigma}$ eliminates spin-up electrons, namely N_{01} and N_{11} continuously decrease, increasing B. (II) The DOS modification which depopulates all excited states, regardless of their spin^{4,7}, namely the eventual decay of N_{10} . Finally, in Figs.2 (d-f), we witness the survival of only N_{00} at very low T, since $U_{o\sigma}$ acquires its bigger value at zero temperature. Increasing T, $U_{o\sigma}$ decreases, augmenting the influence of the spin-up electrons.

Figure 3 depicts the relative influence of the Zeeman term, α , and the exchange term, β , in wide B and T ranges, for a n-doped ZnSe / Zn_{1-x-y}Cd_xMn_ySe /n-doped ZnSe QW with L=60 nm and y=0.035. In reality L is of no importance here due to the negligible impact of the "feedback mechanism" with these material parameters. For comparison we notice that the conduction band-offset, $\Delta U_{cb}=1$ Hartree* ≈ 70.5 meV. The spin splitting in the present article, $U_{o\sigma}=\alpha+\beta$, while $U_{o\sigma}^{@}$ was used in our previous low-T calculations⁷ ($B_{5/2}(\xi)$ approximated by 1, and α ignored). Figure 3 elaborates the competition between B (aligning spins) and T (bringing on anarchy). Figure 3b

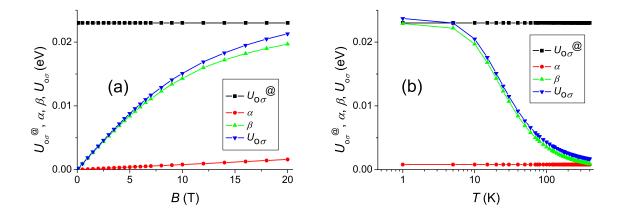


FIG. 3: (Color online) The relative influence of the Zeeman term, α , and the exchange term β in wide B and T ranges, for a n-doped ZnSe / Zn_{1-x-y}Cd_xMn_ySe /n-doped ZnSe QW with L=60 nm and y=0.035. $-J_{sp-d}=12\times 10^{-3}$ eV nm³. Of course, $\alpha=\alpha(B)$, while $\beta=\beta(B,T)$. (a) $\alpha=\alpha(B)$, $\beta=\beta(B)$ for T=20 K. (b) $\alpha=\alpha(T)=constant$, $\beta=\beta(T)$ for B=10 T. L=60 nm. Each panel also contains the spin-splitting, $U_{o\sigma}=\alpha+\beta$, as well as the value of the spin-splitting used in our previous low-T calculation⁷ (i.e. taking into account only β and approximating the corresponding Brillouin function by 1), $U_{o\sigma}^0$. For comparison we notice that the conduction band-offset, $\Delta U_{cb}=1$ Hartree* ≈ 70.5 meV.

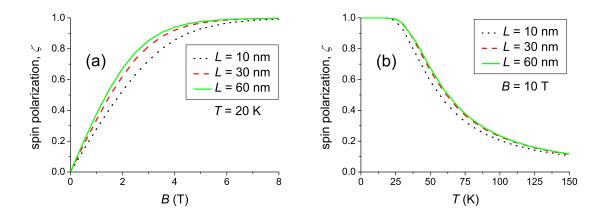


FIG. 4: (Color online) The spin polarization, ζ , tuned by varying: (a) the in-plane magnetic field, B, keeping T=20 K (left panel), and (b) the temperature, T, keeping B=10 T (right panel), for different well widths, L=10 nm, 30 nm, and 60 nm. $-J_{sp-d}=12\times10^{-3}$ eV nm³.

justifies our previous low-temperature approximation: at low enough T, $U_{o\sigma} \approx U_{o\sigma}^{@}$. At higher temperatures, $B_{5/2}(\xi)$ cannot be approximated with 1. As k_BT increases, ξ decreases, and consequently $B_{5/2}(\xi) < 1$. In other words, increasing T, the spin-splitting decreases allowing enhanced contribution of the spin-up electrons to the system's properties. Finally we notice that an opposite sign of g^* (e.g. CdTe vs. ZnSe) is expected to have small effect on the results since the most important term is β .

Figure 4 depicts the spin polarization tuned by varying the parallel magnetic field and the temperature, for different choices of the well width. Since for $B \geq 8$ T, $\zeta = 1$, only the range $B \in [0, 8$ T] is presented in Fig. 4a. Since for $T \geq 150$ K, ζ is less than ≈ 0.1 , only the range $T \in [0, 150$ K] is presented in Fig. 4b. Because of the DOS modification⁷, resulting in different distribution of electrons among the spin-subbands (cf. Fig.2), we observe a clear dependence of $\zeta = \zeta(L)$, i.e. $\zeta(L = 60 \text{ nm}) > \zeta(L = 30 \text{ nm}) > \zeta(L = 10 \text{ nm})$. We also observe that for B = 0, ζ vanishes, i.e. there is no spontaneous spin polarization phase due to the tiny "feedback mechanism" for this choice of material parameters.

Subsequently we deliberately increase $-J_{sp-d}$ by an order of magnitude i.e. we present in Fig. 5 results with $-J_{sp-d}=12\times 10^{-2}$ eV nm³ (which is of a little smaller magnitude than the value commonly used^{19,29,30} for the III-V Ga(Mn)As valence band system, $J_{pd}=15\times 10^{-2}$ eV nm³). L=60 nm and T=20 K. Comparing Fig. 5 with

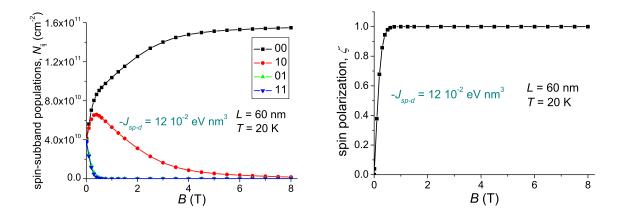


FIG. 5: (Color online) The spin-subband populations, N_{ij} (left panel) and the spin polarization, ζ (right panel) tuned by varying B for L=60 nm, T=20 K, using $-J_{sp-d}=12\times10^{-2}$ eV nm³.

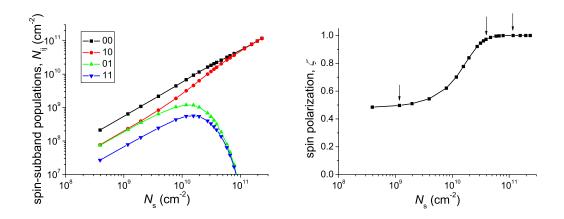


FIG. 6: (Color online) The spin-subband populations, N_{ij} (left panel) and the spin polarization, ζ (right panel), tuned by varying the sheet carrier concentration, N_s , for L=60 nm, T=20 K and B=0.01 T, using $J=12\times10^{-1}$ eV nm³. The little arrows indicate N_s values where we also compare with B=0.0001 T in the text.

Fig. 2c and Fig. 4a we observe that: (α') The greater value of $-J_{sp-d}$ makes it much easier to attain a completely spin-polarized system $(\zeta = 1)$ i.e. for $B \ge 1$ T instead of $B \ge 8$ T. (β') Initially, increasing B, due to the increased $U_{o\sigma}$, N_{10} grows, in contrast to Fig. 2c. Naturally, subsequently N_{10} is depopulated because of the in-plane magnetic field induced DOS modification. (γ') Although the system is more susceptible to spin-polarization, still, practically no spontaneous spin-polarization phase exists for B = 0, at this temperature.

Up to now, we have deliberately kept the total sheet carrier concentration constant. Below we examine the influence of N_s on the spin-subband populations and the spin polarization for different values of the magnitude of the spin-spin exchange interaction, J. Since N_s is affected by many factors (QW profile, material properties, valence-band- or conduction-band-based structures etc) we have decided to use J as a parameter here. Naturally, in a heterostructure where higher N_s can be achieved we may require smaller values of J in order to completely spin-polarize carriers. Using the rest material parameters as above but modifying J, we have systematically studied the N_s influence. For $J=12\times 10^{-2}~{\rm eV}~{\rm nm}^3$ there is a very small influence of N_s on ζ . The situation changes using $J=12\times 10^{-1}~{\rm eV}~{\rm nm}^3$. Figure 6 shows N_{ij} and ζ tuned by varying N_s for $L=60~{\rm nm}$, $T=20~{\rm K}$ and $B=0.01~{\rm T}$, using $J=12\times 10^{-1}~{\rm eV}~{\rm nm}^3$. We observe that increase of N_s from $\approx 1.0\times 10^9~{\rm cm}^{-2}$ to $\approx 1.0\times 10^{11}~{\rm cm}^{-2}$ is sufficient to completely spin-polarize carriers. This is purely due to the "feedback mechanism" stemming from the difference between the populations of spin-down and spin-up carriers. If we decrease B from 0.01 T to 0.0001 T, then e.g. (a) for $N_s=1.175\times 10^9~{\rm cm}^{-2}$, ζ changes from 0.497 to 0.005, (b) for $N_s=3.917\times 10^{10}~{\rm cm}^{-2}$, ζ changes from 0.973 to 0.909, however, (c) for $N_s=1.175\times 10^{11}~{\rm cm}^{-2}$, ζ remains 1.

IV. CONCLUSION

We have studied the spin-subband structure of quasi two-dimensional carriers in dilute-magnetic-semiconductorbased heterostructures, under the influence of an in-plane magnetic field. The proper density of states was used for the first time, incorporating the dependence on the in-plane wave vector perpendicular to the in-plane magnetic field. We have examined the interplay between different degrees of spatial and magnetic confinement, as well as the influence of temperature in a wide range. We have systematically studied the spin-subband populations and the spin-polarization as functions of the temperature and the in-plane magnetic field. We have examined a wide range of material and structural parameters, focusing on the quantum well width, the magnitude of the spin-spin exchange interaction, and the sheet carrier concentration. In particular we have shown that with sufficient magnitude of the spin-spin exchange interaction, the sheet carrier concentration emerges as an important factor to manipulate the spin-polarization, inducing spontaneous (i.e. for $B \to 0$) spin-polarization. We have shown how at low temperatures the spin-splitting acquires its bigger value and how it decreases at higher temperatures. Increasing the in-plane magnetic field, the spin-splitting increases inducing depopulations of the "minority"-spin subbands. Moreover, the DOS modification induces depopulations of all energetically higher subbands. Finally, we have indicated the ranges where the system is completely spin-polarized.

For conduction band electrons, we use $g^* = 1.37$ for ZnSe^{12} , and $g^* = -1.644$ for CdTe^{13} . Some of the values found in the literature are quoted below:

l	g^*	Ref. 12	Ref. 13	Ref. 14	$\mathrm{Ref.}^{15}$
	ZnSe	1.37		1.15	1.7
	CdTe		-1.644	-1.59	-1.1

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